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(54) Title: PROCESS FOR ALKOXYLATION OF DI, TRI AND POLYALCOHOLS

(57) Abstract: A process for alkoxylation of a di, tri or polyalcohol, being solid at applied reaction temperature, in the presence of a catalytically effective amount of at least one alkoxylation catalyst is disclosed. The process is a solvent free process and comprises the steps of i) under effective stirring submitting at least one said di, tri or polyalcohol to reaction with at least one alkylene oxide, said di, tri or polyalcohol being coated with a catalytically effective amount of said at least one alkoxylation catalyst, and ii) under effective stirring charging to in step (i) obtained product and submitting to reaction further amounts of at least one said di, tri or polyalcohol and at least one alkylene oxide, and optionally iii) under effective stirring submitting in step (ii) obtained product to reaction with a further amount of at least one alkylene oxide and/or at least one di, tri or polyalcohol.

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## PROCESS FOR ALKOXYLATION OF DI, TRI AND POLYALCOHOLS

The present invention relates to a novel improved solvent free process for alkoxylation of di, tri and polyalcohols being solid at applied alkoxylation temperature, such as di, tri, and polyalcohols having a melting point of 100°C or more.

Alkoxylated di, tri and polyalcohols are polyethers obtained by polyaddition of alkylene oxides, such as ethylene oxide, propylene oxide, butylene oxide, butadien monoxide, cyclohexene oxide and phenylethylene oxide to alcohols, such as di, tri and polyalcohols, in the presence of an effective amount of an alkoxylation catalyst. Said polyethers are hydroxyfunctional compounds having a functionality supposed to be equivalent to that of said alcohol.

Ethoxylation and propoxylation are the most important alkoxylation. Propoxylation yields mainly secondary hydroxyl groups. An undesired side reaction is parallel formation of unsaturated end groups. Formation of unsaturated end groups accelerates along with increased alkoxylation temperature and increased molecular weight of the polyether produced and results in a decreased polymerisation rate and reduced functionality. Ethoxylation only yields primary hydroxyl groups and the problem with undesired end unsaturation does not exist.

High melting solid di, tri and polyalcohols requires a reduction of the melting point, such as esterification with for instance a fatty acid, and/or a co-solvent, such as organic solvents and/or water, in order to make alkoxylation feasible at temperatures and pressures obtainable in typical alkoxylation plants. This is an important factor distinguishing alkoxylation of high melting solid di, tri and polyalcohols from other categories of substrates typically subjected to alkoxylation.

Water is a frequently used solvent in alkoxylation of solid di, tri or polyalcohols. Whilst the di, tri or polyalcohol must be at least partially dissolved in the water, it is not necessary that the entire amount of said alcohol is in solution. Thus it is possible to employ a suspension of said alcohol in its own aqueous solution. Usually 10 to 50 parts by weight of water are used for each 100 parts of di, tri or polyalcohol. The use of water as solvent leads to undesired formation of glycols and polyglycols from reaction between water and alkylene oxide. This reaction interferes in the process and the product obtained is a mixture of glycols, polyglycols, polymers and oligomers of said alcohol and alkylene oxide, and water, which mixture not is easily separated.

Only a limited amount of organic solvents, such as aromatic hydrocarbons, chlorinated hydrocarbons, oxygenated organic compounds, alkylethers of glycols and saturated

hydrocarbons, being inert to reaction with alkylene oxides, is readily available. Most organic solvents react with the alkylene oxide contaminating obtained products. The use of organic solvents furthermore creates undesired hazards and necessitates refinery of obtained products.

During for instance a normally employed process for ethoxylation of a di, tri or polyalcohols which not is at hand in a liquid state at the reaction temperature, the reaction is carried out with water and/or other polar solvents present so that the reaction takes place in a single phase system. By-products as for instance mono, di and triethylene glycol are during the process formed and are very difficult to remove due to a low volatility, formation of azeotropic mixtures and a strong affinity to the main product. The presence of these ethylene glycols in the ethoxylated di, tri or polyalcohol results in for instance acrylations in formation of corresponding acrylates of mono, di and triethylene glycol, which are toxic and skin irritating. This is valid especially for diethylene glycol diacrylate.

A possible solution to above discussed problems is disclosed in US 2,902,478 teaching that heat sensitive polyols, which are substantially insoluble in alkylene oxides, can be alkoxylation at temperatures below their melting point and decomposition temperatures and in the absence of solvents by the use of trimethylamine as alkoxylation catalyst. A drawback is that amine catalysts inherently are unable to catalyse reactions of alkylene oxides beyond low molecular weights.

It is furthermore known in the art to alkoxylation high melting polyols, such as pentaerythritol, in mixtures with low melting or liquid di or trifunctional alcohols, such as glycols and trimethylolpropane. Obtained products are mixtures of alkoxylation impossible or difficult to separate.

Yet a further known concept for alkoxylation of high melting solid di, tri, and polyalcohols include the use of alkylene carbonates as disclosed in US Patent 2,766,292.

Problems and drawbacks when reacting polyols with alkylene oxides are thoroughly discussed in for instance US Patent 2,766,292.

The present invention provides a novel and improved process for alkoxylation of di, tri and polyalcohols being solid at applied alkoxylation temperature, wherein above disclosed problems and drawbacks, such as formation of glycols and polyglycols by reaction between water and alkylene oxides and/or formation of by-products by reaction between organic solvents and alkylene oxides, are avoided or substantially reduced. The level of undesired glycols, such as mono, di and triethylene glycol is considerably reduced in products obtained

by the process of the present invention compared to products obtained from processes employing a co-solvent, such as water.

The process of the present invention is a substantially solvent, including water, free process performed in the presence of a catalytically effective amount of at least one alkoxylation catalyst. Any water of solvent present in said process amounts to less than 10%, such as less than 5% or less than 1%. Said process comprises:

- i) under effective stirring submitting at least one di, tri or polyalkohol, being solid at applied reaction temperature such as having a melting point of 100°C or more, to reaction with at least one alkylene oxide, said alcohol being coated with a catalytically effective amount of said at least one alkoxylation catalyst and said reaction being performed at a molar ratio said alcohol to said alkylene oxide yielding an alkoxyated alcohol oligomer or polymer (a), which oligomer or polymer is liquid at applied reaction temperature, and
- ii) under effective stirring charging to in Step (i) obtained product and submitting to reaction further amounts of at least one said di, tri or polyalcohol and at least one alkylene oxide, said alcohol and said alkylene oxide being charged at a molar ratio yielding a liquid alkoxyated alcohol oligomer or polymer (b), which oligomer or polymer is liquid at applied reaction temperature, and optionally
- iii) under effective stirring submitting in Step (ii) obtained product to reaction with a further amount of at least one alkylene oxide and/or at least one di, tri or polyalcohol, said alkylene oxide and/or said alcohol being charged in an amount yielding an alkoxyated alcohol oligomer or polymer (c) having at least 0.5 alkoxy units.

Said effective stirring is preferably performed using an agitator having a diameter of at least 90%, such as 90-95% or 96-99%, of the diameter of the reactor. It is important for alkoxylation of di, tri and polyalcohols, being solid at applied reaction temperature, to assure efficient mixing of the reagents, especially during the initial Step (i) of the process of the present invention. An efficient agitator influence the performance of the reaction and distribution of obtained product.

The di, tri or polyalcohol charged in Step (ii) and/or in optional Step (iii) can in the various preferred embodiments of the present invention suitably and individually be coated with a catalytically effective amount of said alkoxylation catalyst. The alkoxylation catalyst can in alternative and preferred embodiments in a catalytically effective amount be charged to the reaction mixture of said Step (ii) and/or said optional Step (iii).

Various embodiments of the process according to the present invention include that the alkoxyated alcohol oligomer or polymer (a) yielded in Step (i) and the alkoxyated alcohol

oligomer or polymer (b) yielded in Step (ii) individually has at least 0.5, such as 0.5-10, 2-5 alkoxy units, and that the alkoxyated alcohol oligomer or polymer (c) yielded in Step (iii) has at least 0.5, such as 0.5-50, 1-30 or 2-20, alkoxy units.

Step (i) of the present process is in a preferred embodiment a single batch Step performed at a molar ratio employed di, tri or polyalcohol to employed alkylene oxide of between 0.5 and 10, such as between 2 and 5. In a likewise preferred embodiment, Step (ii) is a continuous Step performed at a molar ratio employed di, tri or polyalcohol to employed alkylene oxide of between 0.5 and 10, such as between 2 and 5. Said optional Step (iii) is preferably a repeated batch Step performed at a molar ratio yielding an alkoxyated alcohol oligomer or polymer having between 0.5 and 50, such as between 1 and 10, between 2 and 20, between 2 and 30 or between 1 and 30, alkoxy units. Each Step is preferably and individually performed at a temperature of 50-250°C, such as 80-200°C or 120-180°C.

The di, tri or polyalcohol employed in Step (i), Step (ii) and/or optional Step (iii) is in preferred embodiments individually a di, tri or polyalcohol having a melting point of at least 100°C at atmospheric pressure, such as pentaerythritol, dipentaerythritol, tripentaerythritol, trimethylolethane, ditrimethylolethane, ditrimethylolpropane, sorbitol or sucrose. Said di, tri or polyalcohol is most preferably pentaerythritol or dipentaerythritol.

The alkylene oxide employed in Step (i), Step (ii) and/or optional Step (iii) is in preferred embodiments of the present invention individually ethylene oxide, propylene oxide, butylene oxide, butadiene monoxide, cyclohexene oxide and/or phenylethylene oxide and most preferably ethylene oxide, propylene oxide or a mixture or combination thereof.

Preferred alkoxylation catalysts include embodiments selected among organic acidic compounds and organic or inorganic alkaline compound. The alkylation catalyst employed in Step (i), Step (ii) and optional Step (iii) is most preferably and individually at least one alkali metal hydroxide or alkoxide, such as methoxide, and/or alkaline earth metal hydroxide. Suitable alkoxylation catalysts can be exemplified by compounds such as sodium hydroxide, sodium methoxide, potassium hydroxide, potassium methoxide, lithium hydroxide and lithium methoxide. Said alkoxylation catalyst is preferably and individually present in said Step (i), Step (ii) and optional Step (iii) in an amount of 0.001-0.1%, such as 0.001-0.05% or 0.001-0.005%, calculated on final product yielded from the process of the present invention.

The most preferred embodiments of the process according to the present invention include embodiments wherein the di, tri or polyalcohol employed in said Step (i), Step (ii) and optional Step (iii) is pentaerythritol or dipentaerythritol and wherein the alkylene oxide employed in said Steps is ethylene oxide and/or propylene oxide.

Without further elaboration, it is believed that one skilled in the art can, using the preceding description, utilise the present invention to its fullest extent. The following preferred specific embodiments are, therefore, to be construed as merely illustrative and not limitative of the remainder of the disclosure in any way whatsoever. In the following Examples 1 and 2 refer to coating of pentaerythritol and dipentaerythritol with an alkoxylation catalyst, Examples 3-6 disclose embodiments, of the process of the present invention, wherein pentaerythritol and dipentaerythritol are ethoxylated at various ethoxylation degrees and Example 7 is a comparative example wherein pentaerythritol is ethoxylated, in the presence of water as co-solvent, in accordance with a commercially known and available process.

### Example 1

Coating of pentaerythritol with potassium hydroxide (alkoxylation catalyst).

300 ml of an aqueous potassium hydroxide solution was prepared at a concentration equivalent to 0.002% KOH on final alkoxyated product. 300 g of pentaerythritol was added to said solution and the mixture was homogenised. Water was subsequently evaporated by drying at 120°C. The resulting product was finally grinded to a powder.

### Example 2

Coating of dipentaerythritol with potassium hydroxide (alkoxylation catalyst).

600 ml of an aqueous potassium hydroxide solution was prepared at a concentration equivalent to 0.002% KOH on final alkoxyated product. 600 g of dipentaerythritol was added to said solution and the mixture was homogenised. Water was subsequently evaporated by drying at 120°C. The resulting product was finally grinded to a powder.

### Example 3

Preparation in 2 steps of ethoxylated pentaerythritol having an average ethoxylation degree of 5 moles of ethylene oxide / mole of pentaerythritol using 0.002% KOH on final product as alkoxylation catalyst.

#### Step (i)

200 g of pentaerythritol coated with KOH in accordance with Example 1 was weighed into an adequately equipped (including an agitator having a diameter of 90-95% of the autoclave

diameter) 2 litres laboratory autoclave. 131 g of ethylene oxide was under efficient stirring during 2 hours charged to and reacted with the pentaerythritol at a temperature of 160-180°C. The pressure during said reaction was 200-600 kPa.

Determined ethoxylation degree*	2.04
Yield, %	99.7

#### Step (ii)

18.2 g of the product obtained in Step (i) was weighed into a 2 litres laboratory autoclave equipped as in Step (i) and mixed with 0.98 g of an aqueous KOH solution (1.04 g KOH/100 ml) as alkoxylation catalyst. 310 g of ethylene oxide and 181.8 g of pentaerythritol were under efficient stirring during 8 hours charged to said Step (i) product and submitted to reaction at a temperature of 160-180°C. The pressure during said reaction was 200-600 kPa.

Determined ethoxylation degree*	5.12
Yield, %	94.9

The content of monoethylene, diethylene and triethylene glycol in the final product is given in Table 1.

\* Moles of ethylene oxide / mole of pentaerythritol.

#### **Example 4**

Preparation in 3 steps of ethoxylated pentaerythritol having an average ethoxylation degree of 5 moles of ethylene oxide / mole of pentaerythritol using 0.002% KOH on final product as alkoxylation catalyst.

#### Step (i)

200 g of pentaerythritol coated with KOH in accordance with Example 1 was weighed into an adequately equipped (including an agitator having a diameter of 90-95% of the autoclave diameter) 2 litres laboratory autoclave. 131 g of ethylene oxide was under efficient stirring during 2 hours charged to and reacted with the pentaerythritol at a temperature of 160-180°C. The pressure during said reaction was 200-600 kPa.

Determined ethoxylation degree*	2.04
Yield, %	99.7

Step (ii)

18.2 g of the product obtained in Step (i) was weighed into a 2 litres laboratory autoclave equipped as in Step (i) and mixed with 0.61 g of an aqueous KOH solution (1.04 g KOH/100 ml) as alkoxylation catalyst. 126 g of ethylene oxide and 181.8 g of pentaerythritol were under efficient stirring during 1.5 hour charged to said Step (i) product and submitted to reaction at a temperature of 160-180°C. The pressure during said reaction was 200-600 kPa.

Determined ethoxylation degree*	2.15
Yield, %	99.5

Step (iii)

199.4 g of the product obtained in Step (ii) was weighed into a 2 litres laboratory autoclave equipped as in Step (i) and Step (ii) and mixed with 0.37 g an aqueous KOH solution (1.04 g KOH/100 ml) as alkoxylation catalyst. 170 g of ethylene oxide was under stirring during 2.5 hours charged to said Step (ii) product and submitted to reaction at a temperature of 160-180°C. The pressure during said reaction was 200-600 kPa.

Determined ethoxylation degree*	5.12
Yield, %	94.9

The content of monoethylene, diethylene and triethylene glycol in the final product is given in Table 1.

\* Moles of ethylene oxide / mole of pentaerythritol.

**Example 5**

Preparation in 2 steps of ethoxylated dipentaerythritol having an average ethoxylation degree of 13 moles of ethylene oxide / mole of dipentaerythritol using 0.002% KOH on final product as alkoxylation catalyst.

Step (i)

293 g of dipentaerythritol coated with KOH in accordance with Example 2 was weighed into an adequately equipped (including an agitator having a diameter of 90-95% of the autoclave diameter) 2 litres laboratory autoclave. 259 g of ethylene oxide was under efficient stirring



during 3 hours charged to and reacted with the dipentaerythritol at a temperature of 160-180°C. The pressure during said reaction was 200-600 kPa.

Determined ethoxylation degree*	5.0
Yield, %	98.7

#### Step (ii)

23 g of the product obtained in Step (i) was weighed into a 2 litres laboratory autoclave equipped as in Step (i) and mixed with 1.52 g of an aqueous KOH solution (1.04 g KOH/100 ml) as alkoxylation catalyst. 534 g of ethylene oxide and 230 g of dipentaerythritol were under efficient stirring during 3 hours charged to said Step (i) product and submitted to reaction at a temperature of 160-180°C. The pressure during said reaction was 200-600 kPa.

Determined ethoxylation degree*	13.03
Yield, %	97.2

The content of monoethylene, diethylene and triethylene glycol in the final product is given in Table 1.

\* Moles of ethylene oxide / mole of dipentaerythritol.

#### **Example 6**

Preparation in 3 steps of ethoxylated dipentaerythritol having an average ethoxylation degree of 9 moles of ethylene oxide / mole of dipentaerythritol using 0.002% KOH on final product as alkoxylation catalyst.

#### Step (i)

293 g of dipentaerythritol coated with KOH in accordance with Example 2 was weighed into an adequately equipped (including an agitator having a diameter of 90-95% of the autoclave diameter) 2 litres laboratory autoclave. 259 g of ethylene oxide was under efficient stirring during 3 hours charged to and reacted with the dipentaerythritol at a temperature of 160-180°C. The pressure during said reaction was 200-600 kPa.

Determined ethoxylation degree*	5.0
Yield, %	98.7

Step (ii)

18.2 g of the product obtained in Step (i) was weighed into a 2 litres laboratory autoclave equipped as in Step (i) and mixed with 0.67 g of an aqueous KOH solution (1.04 g KOH/100 ml) as alkoxylation catalyst. 158 g of ethylene oxide and 181.8 g of dipentaerythritol were under efficient stirring during 3.5 hours charged to said Step (i) product and submitted to reaction at a temperature of 160-180°C. The pressure during said reaction was 200-600 kPa.

Determined ethoxylation degree*	4.8
Yield, %	97.6

Step (iii)

357.4 g of the product obtained in Step (ii) was weighed into a 2 litres laboratory autoclave equipped as in Step (i) and Step (ii) and mixed with 0.28 g an aqueous KOH solution (1.04 g KOH/100 ml) as alkoxylation catalyst. 140 g of ethylene oxide was under stirring during 4.5 hours charged to said Step (ii) product and submitted to reaction at a temperature of 160-180°C. The pressure during said reaction was 200-600 kPa.

Determined ethoxylation degree*	9.02
Yield, %	93.2

The content of monoethylene, diethylene and triethylene glycol in the final product is given in Table 1.

\* Moles of ethylene oxide / mole of dipentaerythritol.

**Example 7**

Preparation in 2 steps of ethoxylated dipentaerythritol having an average ethoxylation degree of 5 moles of ethylene oxide / mole of dipentaerythritol using 0.002% KOH on final product as alkoxylation catalyst.

Step (i)

293 g of dipentaerythritol coated with KOH in accordance with Example 2 was weighed into an adequately equipped (including an agitator having a diameter of 90-95% of the autoclave diameter) 2 litres laboratory autoclave. 260 g of ethylene oxide was under efficient stirring

during 3.5 hours charged to and reacted with the dipentaerythritol at a temperature of 160-180°C. The pressure during said reaction was 200-600 kPa.

Determined ethoxylation degree*	4.9
Yield, %	97.7

#### Step (ii)

18.2 g of the product obtained in Step (i) was weighed into a 2 litres laboratory autoclave equipped as in Step (i). 162 g of ethylene oxide and 181.8 g of dipentaerythritol coated with KOH in accordance with Example 2 were under efficient stirring during 4.5 hours charged to said Step (i) product and submitted to reaction at a temperature of 160-180°C. The pressure during said reaction was 200-600 kPa.

Determined ethoxylation degree*	5.1.
Yield, %	99.3

The content of monoethylene, diethylene and triethylene glycol in the final product is given in Table 1.

\* Moles of ethylene oxide / mole of dipentaerythritol.

#### **Example 7 - Comparative**

Preparation ethoxylated pentaerythritol having an average ethoxylation degree of 5 moles ethylene oxide / mole of pentaerythritol using 0.1% of KOH on final product as alkoxylation catalyst and water as co-solvent.

136 g of pentaerythritol, 3.6 g of powdered KOH and 70 g of distilled water were weighed into an adequately equipped 1 litre laboratory autoclave. The mixture was while stirring heated to 150°C under nitrogen atmosphere. 220 g of ethylene oxide was then under stirring and during 3.5 hours charged to obtained mixture and submitted to reaction at a temperature of 150°C and at a pressure of 2000-4000 mm Hg.

**Table 1**

	Example 3	Example 4	Example 5	Example 6	Example 7 Comp.
Monoethylene glycol, %	< 0.1	0.1	< 0.1	< 0.1	0.1
Diethylene glycol, %	0.1	0.1	0.3	0.2	0.9
Triethylene glycol, %	0.2	0.2	0.5	0.4	2.6

As can be seen from Table 1, the amount of glycols, especially the amount of di and triglycols, formed from the alkylene oxide is substantially reduced in products obtained by the process of the present invention.

## CLAIMS

1. A process for alkoxylation of a di, tri or polyalcohol, being solid at applied reaction temperature, in presence of a catalytically effective amount of at least one alkoxylation catalyst characterised in, that said process is a substantially solvent, including water, free process comprising the Steps of
  - i) under effective stirring submitting at least one said di, tri or polyalcohol to reaction with at least one alkylene oxide, said alcohol being coated with a catalytically effective amount of said at least one alkoxylation catalyst and said reaction being performed at a molar ratio said alcohol to said alkylene oxide yielding an alkoxyated alcohol oligomer or polymer (a) being liquid at applied reaction temperature, and
  - ii) under effective stirring charging to in Step (i) obtained product and submitting to reaction further amounts of at least one said di, tri or polyalcohol and at least one alkylene oxide, said di, tri or polyalcohol and said alkylene oxide being charged at a molar ratio yielding an alkoxyated alcohol oligomer or polymer (b) being liquid at applied reaction temperature, and optionally
  - iii) under effective stirring submitting in Step (ii) obtained product to reaction with a further amount of at least one alkylene oxide and/or at least one di, tri or polyalcohol, said alkylene oxide and/or said di, tri or polyalcohol being charged in an amount yielding an alkoxyated alcohol oligomer or polymer (c) having at least 0.5 alkoxy units.
2. A process according to Claim 1 characterised in, that said effective stirring is performed using an agitator having a diameter of at least 90% of corresponding reactor diameter.
3. A process according to Claim 1 or 2 characterised in, that the di, tri or polyalcohol charged in Step (ii) is coated with a catalytically effective amount of said at least one alkoxylation catalyst.
4. A process according to Claim 1 or 2 characterised in, that a catalytically effective amount of said alkoxylation catalyst is charged in said Step (ii).
5. A process according to any of the Claims 1-4 characterised in, that the di, tri or polyalcohol charged in said optional Step (iii) is coated with a catalytically effective amount of said at least one alkoxylation catalyst.

6. A process according to any of the Claims 1-4 characterised in, that a catalytically effective amount of said alkoxylation catalyst is charged in said optional Step (iii).
7. A process according to any of the Claims 1-6 characterised in, that said alkoxyated alcohol oligomer or polymer (a) has at least 0.5 alkoxy units
8. A process according to any of the Claims 1-7 characterised in, that said alkoxyated alcohol oligomer or polymer (b) has at least 0.5 alkoxy units.
9. A process according to any of the Claims 1-8 characterised in, that said alkoxyated alcohol oligomer or polymer (c) has at least 0.5 alkoxy units.
10. A process according to any of the Claims 1-9 characterised in, that Step (i) is a single batch step performed at a molar ratio employed di, tri or polyalcohol to employed alkylene oxide of between 0.5 and 10, inclusive.
11. A process according to any of the Claims 1-10 characterised in, that Step (i) is performed at a molar ratio employed di, tri or polyalcohol to employed alkylene oxide of between 2 and 5, inclusive.
12. A process according to any of the Claims 1-11 characterised in, that Step (ii) is a continuous Step performed at a molar ratio employed di, tri or polyalcohol to employed alkylene oxide of between 0.5 and 10, inclusive.
13. A process according to any of the Claims 1-12 characterised in, that Step (ii) is performed at a molar ratio employed di, tri or polyalcohol to employed alkylene oxide of between 2 and 5, inclusive.
14. A process according to any of the Claims 1-13 characterised in, that optional Step (iii) is a batch Step performed at a molar ratio yielding an alkoxyated alcohol oligomer or polymer having between 0.5 and 50 alkoxy units.
15. A process according to any of the Claims 1-14 characterised in, that optional Step (iii) is a batch Step performed at a molar ratio yielding an alkoxyated alcohol oligomer or polymer having between 2 and 20 alkoxy units.
16. A process according to any of the Claims 1-15 characterised in, that Step (i), Step (ii) and/or optional Step (iii) individually is performed at a temperature of 50-250°C.

17. A process according to any of the Claims 1-16 characterised in, that Step (i), Step (ii) and/or optional Step (iii) individually is performed at a temperature of 80-200°C.
18. A process according to any of the Claims 1-17 characterised in, that Step (i), Step (ii) and/or optional Step (iii) individually is performed at a temperature of 120-180°C.
19. A process according to any of the Claims 1-18 characterised in, that the di, tri or polyalcohol employed in Step (i) has a melting point of at least 100°C at atmospheric pressure.
20. A process according to any of the Claims 1-19 characterised in, that the di, tri or polyalcohol employed in said Step (i), Step (ii) and optional Step (iii) individually is pentaerythritol, dipentaerythritol, tripentaerythritol, trimethylolethane, ditrimethylolethane, ditrimethylolpropane, sorbitol or sucrose.
21. A process according to any of the Claims 1-20 characterised in, that the alkylene oxide employed in said Step (i), Step (ii) and optional Step (iii) individually is ethylene oxide, propylene oxide, butylene oxide, butadiene monoxide, cyclohexene oxide and/or phenylethylene oxide.
22. A process according to any of the Claims 1-21 characterised in, that employed alkoxylation catalyst is at least one organic acidic compound or at least one organic or inorganic alkaline compound.
23. A process according to any of the Claims 1-22 characterised in, that employed alkoxylation catalyst is at least one alkali metal hydroxide, alkali metal methoxide or alkaline earth metal hydroxide.
24. A process according to any of the Claims 1-23 characterised in, that the alkoxylation catalyst employed in said Step (i), Step (ii) and optional Step (iii) individually is sodium hydroxide or methoxide, potassium hydroxide or methoxide and/or lithium hydroxide or methoxide.
25. A process according to any of the Claims 1-23 characterised in, that said alkoxylation catalyst in said Step (i), Step (ii) and optional Step (iii) individually is present in an amount of 0.001-0.1% calculated on final product.
26. A process according to any of the Claims 1-25 characterised in, that said alkoxylation catalyst in said Step (i), Step (ii) and optional Step (iii) individually is present in an amount of 0.001-0.05% calculated on final product.

27. A process according to any of the Claims 1-26 characterised in, that said alkoxylation catalyst in said Step (i), Step (ii) and optional Step (iii) individually is present in an amount of 0.001-0.005% calculated on final product.
28. A process according to any of the Claims 1-27 characterised in, that the di, tri or polyalcohol employed in said Step (i), Step (ii) and optional Step (iii) is pentaerythritol or dipentaerythritol.
29. A process according to any of the Claims 1-28 characterised in, that the alkylene oxide employed in Step (i), Step (ii) and optional Step (iii) is ethylene oxide and/or propylene oxide.
30. A process according to any of the Claims 1-29 characterised in, that any water of solvent present in said process amounts to less than 10%, such as less than 5% or less than 1%.



## INTERNATIONAL SEARCH REPORT

International application No.

PCT/SE 02/01707

## A. CLASSIFICATION OF SUBJECT MATTER

IPC7: C07C 41/03, C07C 27/00, B01J 23/02

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC7: C07C, B01J

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

SE,DK,FI,NO classes as above

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

EPO INTERNAL, PAJ, WPI DATA, STN

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 5625045 A (GUPTA ET AL), 29 April 1997 (29.04.97), column 2, line 16 - line 58; column 3, line 47 - column 4, line 2, claims 1-9, ex. 1-5 --	1-30
A	WO 8602635 A1 (THE UNIVER OF MANCHESTER INS OF SCIENCE AND TECHN), 9 May 1986 (09.05.86), page 1, line 3 - line 17; page 2, line 4 - page 3, line 2; page 3, line 6 - page 5, line 2, claims 1-9 --	1-30

☒ Further documents are listed in the continuation of Box C.☒ See patent family annex.

\* Special categories of cited documents:

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier application or patent but published on or after the international filing date

"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance: the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance: the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art

"&amp;" document member of the same patent family

Date of the actual completion of the international search

17 December 2002

Date of mailing of the international search report

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## INTERNATIONAL SEARCH REPORT

International application No.

PCT/SE 02/01707

## C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

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A	EP 0373860 A2 (UNIVER OF MANCHESTER INS OF SCIENCE AND TECHN), 20 June 1990 (20.06.90), column 1, line 1 - line 16; column 1, line 31 - line 53; column 2, line 15 - line 53, column 3, line 17 - line 32; column 3, line 48 - line 51; column 4, line 17 - line 26, claims 1-24 --	1-30
A	US 3085085 A (MARCO WISMER ET AL), 9 April 1963 (09.04.63), column 2, line 22 - line 27; column 3, line 51 - column 4, line 10, claims 1-6 -- -----	1-30

## INTERNATIONAL SEARCH REPORT

01/12/02

International application No.

PCT/SE 02/01707

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